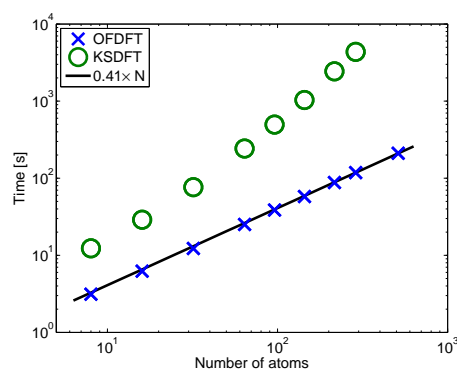


Orbital-free density functional theory implementation within the projector-augmented wave method

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Density Functional Theory (DFT) is the preferred method for studying quantum electronic properties of nanosystems. Its time-dependent extension has allowed simulations to study also excited and dynamical processes. In-depth simulations can be carried on, presently, on systems containing up to 1.000 atoms and process taking up to picoseconds of time. Several orders of magnitude however, are separating these limits to the resources needed to study, for example, nanosystems in aqueous environment, biomolecules and its fluorescence properties. In these important cases, systems can reach up to 10.000 atoms and time scales are of the order nanoseconds. Orbital-Free Density Functional Theory method (OFDFT) can contribute to increase the accessible time and size scales of quantum simulations by several orders of magnitude.

I will present a novel computational scheme of OFDFT method that preserves the promised linear scaling of the method, has good numerical stability and opens the possibility to calculate accurate OFDFT all-electron energy values for any system [1]. Using PAW and real-space methods, our orbital-free results agree with the reference all-electron values with a mean absolute error of 10 meV and the number of iterations required by the self-consistent cycle is comparable to the KS method. The comparison of all-electron and pseudopotential bulk modulus and lattice constant reveal an enormous difference, demonstrating that in order to assess the performance of OFDFT functionals it is necessary to use implementations that obtain all-electron values. The proposed combination of methods is the most promising route currently available. I will finally show an application of the method to the study of parametrized kinetic energy functionals performance and review possible routes in the search of transferable and accurate OFDFT functionals.



References

- [1] J. Lehtomäki, I. Makkonen, M. A. Caro, A. Harju, and O. Lopez-Acevedo, *J. Chem. Phys.* 141, 23, 1 (2014) Preprint available from arXiv:1408.4701.